

## RESEARCH ARTICLE

## Exact response functions within the time-dependent Gutzwiller approach

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We investigate the applicability of the two existing versions of a time-dependent Gutzwiller approach (TDGA) beyond the frequently used limit of infinite spatial dimensions. To this end, we study the two-particle response functions of a two-site Hubbard model where we can compare the exact results and those derived from the TDGA. It turns out that only the more recently introduced version of the TDGA can be combined with a diagrammatic approach which allows for the evaluation of Gutzwiller wave functions in finite dimensions. For this TDGA method we derive the time-dependent Lagrangian for general single-band Hubbard models.

## 1. Introduction

The discovery of high-temperature superconductivity in LaBaCuO by Bednorz and Müller [1] has led to an enormous amount of theoretical and experimental work on unconventional superconductivity (SC) in the past 25 years. There is now a wide agreement that the Coulomb interaction among the conduction electrons of such systems plays an important, or even the main role, in determining the properties of the SC order. Unlike the conventional, i.e., phonon-mediated, pairing which can be understood already on the level of a mean-field theory, a proper treatment of the Coulomb interaction requires genuine many-particle methods. Therefore, our theoretical understanding of correlation-induced superconductivity is still far from satisfactory.

For the theoretical investigation of cuprates, such as LaBaCuO, one often considers a two-dimensional (single-band) Hubbard model

$$\hat{H} = \sum_{i \neq j} \sum_{\sigma=\uparrow, \downarrow} t_{i,j} \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma} + U \sum_i \hat{d}_i \quad \text{with} \quad \hat{d}_i = \hat{n}_{i,\uparrow} \hat{n}_{i,\downarrow} \quad (1)$$

which describes the hopping of electrons with spin  $\sigma$  between lattice sites  $i$  and  $j$ , and  $U\hat{d}_i$  is the Coulomb interaction of electrons on the same site. Describing the superconducting ground states of this (still relatively simple) model, however, is already a challenging task. We have recently introduced a variational approach, based on Gutzwiller wave functions, which allows us to investigate the stability of such states [2, 3]. Unlike alternative methods which are based on the investigation of finite (usually small) clusters, our method addresses the infinite size system.

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This is important because, for an accurate description of superconductivity, one needs a sufficient resolution around the Fermi surface in momentum space.

The Gutzwiller wave function provides an approximation of the many-particle ground state and its properties. In order to go beyond the ground-state description there are two different formalisms which have been proposed in the literature. In Ref. [4], Seibold and Lorenzana introduced a time-dependent Gutzwiller approach (TDGA) which was based on the assumption that the considered frequencies are small compared to typical atomic energies ('antiadiabaticity assumption'). In a number of subsequent works on single-band models [4–14] and (more recently) multi-band models [15, 16] it has been demonstrated that this method provides a much more accurate description of low-energy excitations than, e.g., a mean-field RPA calculation. Based on ideas by Schiro and Fabrizio [17, 18], we have recently derived an improved formulation of the time-dependent Gutzwiller approach which avoids the antiadiabaticity assumption. This method is therefore expected to extend the range of validity of the TDGA to higher frequencies. [19, 20].

In all applications of the TDGA so far, expectation values have been evaluated by means of the 'Gutzwiller approximation' (GA), see below. Certain phenomena, however, cannot be understood within the GA, e.g., the superconductivity in a two-dimensional Hubbard model. Therefore, the main question, which we will address in this work is how the TDGA can be used beyond this approximation, especially by means of the diagrammatic approach which was introduced in Ref. [2].

Our work is organized as follows. In Sec. 2 we introduce the Gutzwiller wave function for the investigation of Hubbard models. The main ideas of the two TDGA formulations are summarised in Sec. 3. To assess the applicability of these TDGA methods beyond the GA, it is instructive to consider a two-site Hubbard model where all expectation values for Gutzwiller wave functions are known exactly. We introduce this model and useful notations in Sec. 4 and investigate its two-particle properties by means of the TDGA in Sec. 5. Finally, in Sec. 6 we formulate the TDGA for lattice systems based on the diagrammatic approach introduced in Ref. [2]. A summary and an outlook close our presentation in Sec. 7.

## 2. Gutzwiller variational wave functions

Gutzwiller wave functions (GWF) allow us to investigate the properties of multi-band Hubbard models. They are defined as [21, 22]

$$|\Psi_G\rangle = \hat{P}_G |\Psi_0\rangle = \prod_i \hat{P}_i |\Psi_0\rangle, \quad (2)$$

where  $|\Psi_0\rangle$  is an arbitrary normalised single-particle product state of the system. The form of the local Gutzwiller correlator depends on the model one aims to investigate. In this work, we will only consider single-band Hubbard models of the form (1). To study such models, Gutzwiller worked with the local correlation operator

$$\hat{P}_i = 1 - (1 - g_i) \hat{d}_i \quad (3)$$

with variational parameters  $g_i$  which connect the non-interacting ( $g_i = 1$ ) and the 'atomic' limit ( $g_i \rightarrow 0$ ) [21]. An alternative definition of the local correlation

operator is given by [22]

$$\hat{P}_i = \sum_{\Gamma} \lambda_{i,\Gamma} |\Gamma\rangle_i \langle \Gamma| , \quad (4)$$

where we introduced the four local basis states  $|\emptyset\rangle_i$ ,  $|\sigma\rangle_i$ ,  $|d\rangle_i$  and corresponding variational parameters  $\lambda_{i,\Gamma}$ . As we have shown in Ref. [2], a diagrammatic evaluation of expectation values for Gutzwiller wave functions is significantly simplified if we write the four parameters  $\lambda_{i,\Gamma}$  as

$$\lambda_{i,\emptyset} = \sqrt{1 + n_{i,\uparrow}^0 n_{i,\downarrow}^0} x_i , \quad \lambda_{i,\sigma} = \sqrt{1 - \bar{n}_{i,\sigma}^0 n_{i,\bar{\sigma}}^0} x_i , \quad \lambda_{i,d} = \sqrt{1 + \bar{n}_{i,\uparrow}^0 \bar{n}_{i,\downarrow}^0} x_i , \quad (5)$$

where the notations  $\bar{\uparrow} = \downarrow$ ,  $\bar{\downarrow} = \uparrow$ ,  $n_{i,\sigma}^0 = \langle \hat{n}_{i,\sigma} \rangle_{\Psi_0}$ ,  $\bar{n}_{i,\sigma}^0 \equiv 1 - n_{i,\sigma}^0$  and the variational parameters  $x_i$  have been introduced.

Despite the different numbers of variational parameters, the correlation operators (3) and (4) (or (4) together with the transformation (5)) lead to the same variational ground state as long as we consider the single-particle state  $|\Psi_0\rangle$  as a variational object. One has to be more cautious, however, about the proper choice of the correlation operator within the time-dependent Gutzwiller theory, as we will discuss in Sec. 5. A generalisation of the Gutzwiller Ansatz for multi-band models is straightforward [22–24] but needs not to be discussed for our considerations in this work.

The evaluation of expectation values for Gutzwiller wave functions is a difficult many-particle problem which can be solved exactly only in a few cases. For a one-dimensional (single-band) model, expectation values have been calculated for homogeneous para- and ferromagnetic states [25, 26]. In the opposite limit of infinite spatial dimensions it is possible to evaluate expectation values for general multi-band models. The energy functional which arises in this limit is usually denoted as the GA when it is applied to finite-dimensional systems. The diagrammatic approach introduced in Ref. [2] provides a systematic way to improve the GA.

### 3. The time-dependent Gutzwiller approach (TDGA)

In this section, we briefly summarise the two ways of formulating a TDGA. The first, which in the following we term the ‘low frequency approximation’ (LFA), has been successfully used previously within the GA. The second is the fully time-dependent approach (FTDA) and will turn out to be the appropriate method for generalizing the calculation beyond the limit of infinite spatial dimensions.

#### 3.1. LFA: the low-frequency approximation

The expectation value of the Hamiltonian with respect to our Gutzwiller wave functions,

$$\langle \hat{H} \rangle_{\Psi_G} = E_G(\{z_i\}, \tilde{\rho}) , \quad (6)$$

is a function of the (non-interacting) density matrix  $\tilde{\rho}$  with the elements

$$\rho_{(j\sigma'),(i\sigma)} = \langle \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma'} \rangle_{\Psi_0} \quad (7)$$

and of the local variational parameters  $z_i$  (i.e.,  $g_i$ ,  $\lambda_{i,\Gamma}$ , or  $x_i$  in the single-band case). We define the effective energy function

$$E(\tilde{\rho}) \equiv \min_{\{z_i\}} E_G(\{z_i\}, \tilde{\rho}) \quad (8)$$

which, like the corresponding Hartree–Fock energy, only depends on the (non-interacting) density matrix  $\tilde{\rho}$ .

For the study of response functions we add a time-dependent field

$$\hat{V}(t) = \sum_{i,j,\sigma} f_{(i\sigma),(j\sigma')}(t) \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma'} \quad (9)$$

to the Hamiltonian  $\hat{H}$  of our system. In the time-dependent Hartree–Fock approximation (also denoted as the ‘random-phase approximation’) the time dependence of  $\tilde{\rho}$  is given by the equation of motion

$$i\dot{\tilde{\rho}}(t) = [\tilde{h}(\tilde{\rho}(t)) + \tilde{f}(t), \tilde{\rho}(t)] \quad (10)$$

where the elements of the Hamilton matrix  $\tilde{h}$  are given by the derivative of the Hartree–Fock energy with respect to the matrix elements of  $\tilde{\rho}$ . It is the main idea of the time-dependent Gutzwiller approach, as introduced in Ref. [4], to evaluate Eq. (10) with a Hamilton matrix  $\tilde{h}$  which is derived from the effective energy function (8),

$$h_{(i\sigma),(j\sigma')} = \frac{\partial}{\partial \rho_{(j\sigma'),(i\sigma)}} E(\tilde{\rho}) . \quad (11)$$

The use of the effective energy function in Eqs. (8), (10) has been denoted as the ‘antiadiabaticity assumption’ in previous works. Physically, it is based on the idea that the time scale of ‘atomic’ fluctuations, described by the parameters  $z_i$ , will be short compared to the externally induced fluctuations of  $\tilde{\rho}(t)$ . Obviously, this approximation is justifiable only for the study of low-frequency excitations.

Within the GA, the further evaluation of Eqs. (8), (10), (11) for the calculation of two-particle response functions has been discussed in great detail in previous work [4–14] and shall not be repeated here. We will analyse these equations for the two-site Hubbard model in Sec. 5.1.

### 3.2. FTDA: The fully time dependent approach

The Schrödinger equation for a general time-dependent Hamiltonian  $\hat{H}(t)$  ( $\hbar = 1$ ), can be obtained by requesting that the action

$$S = \int dt L(t) \quad (12)$$

is stationary with respect to variations of the wave function. It is usually convenient to perform this variation based on a real Lagrangian [27]

$$L(t) = \frac{i}{2} \frac{\langle \Psi | \dot{\Psi} \rangle - \langle \dot{\Psi} | \Psi \rangle}{\langle \Psi | \Psi \rangle} - \frac{\langle \Psi | \hat{H} | \Psi \rangle}{\langle \Psi | \Psi \rangle} \equiv L^{(1)} + L^{(2)} . \quad (13)$$

If one restricts the wave-function  $|\Psi(\{z_i\}, t)\rangle$  to a certain trial form, depending on (in general complex) functions  $z_i$ , the differential equations

$$\frac{d}{dt} \frac{\partial L}{\partial \dot{z}_i^{(*)}} - \frac{\partial L}{\partial z_i^{(*)}} = 0 \quad (14)$$

provide an approximation for the exact time evolution. Note that the exact solution is reproduced by solving Eqs. (14) if the former can be written in the form of the Ansatz wave-function  $|\Psi(\{z\}, t)\rangle$ . Moreover, the Ritz' variational principle is recovered if the Hamiltonian and, consequently, the parameters  $z_i$  are time independent.

Within the time-dependent Gutzwiller theory our obvious choice for an Ansatz wave function is of the form (2) where both,  $|\Psi_0\rangle$  and  $\hat{P}_G$ , may be time dependent. The evaluation of the differential equations (14) then requires the calculation of

$$\frac{\langle \Psi_G | \hat{H} | \Psi_G \rangle}{\langle \Psi_G | \Psi_G \rangle} \quad \text{and} \quad \frac{\langle \Psi_G | \dot{\Psi}_G \rangle}{\langle \Psi_G | \Psi_G \rangle} \quad (15)$$

which, again, constitutes a difficult many-particle problem that cannot be solved in general. In Ref. [19], we have derived the differential equations (14) for general multi-band models by evaluating (15) in the limit of infinite spatial dimensions. The exact evaluation of Eqs. (14) for the case of a two-site Hubbard model will be discussed in Sec. 5.2. Finally, in Sec. 6 we derive the differential equations (14) for general Hubbard models based on the diagrammatic approach introduced in Ref. [2].

#### 4. The two-site Hubbard model

A general two-electron state in the ( $S_z = 0$ ) Hilbert space of the two-site Hubbard model ( $t \geq 0$ )

$$\hat{H}_{2s} = -t \sum_{\sigma} \left( \hat{c}_{1,\sigma}^{\dagger} \hat{c}_{2,\sigma} + \hat{c}_{2,\sigma}^{\dagger} \hat{c}_{1,\sigma} \right) + U \sum_{i=1}^2 \hat{n}_{i,\uparrow} \hat{n}_{i,\downarrow} \quad (16)$$

has the form

$$|\Phi\rangle = \alpha_1 |d, \emptyset\rangle + \alpha_2 |\emptyset, d\rangle + \alpha_3 |\uparrow, \downarrow\rangle + \alpha_4 |\downarrow, \uparrow\rangle \quad (17)$$

where we introduced the four basis states

$$|d, \emptyset\rangle \equiv \hat{c}_{1,\uparrow}^{\dagger} \hat{c}_{1,\downarrow}^{\dagger} |0\rangle, \quad |\emptyset, d\rangle \equiv \hat{c}_{2,\uparrow}^{\dagger} \hat{c}_{2,\downarrow}^{\dagger} |0\rangle, \quad |\uparrow, \downarrow\rangle = \hat{c}_{1,\uparrow}^{\dagger} \hat{c}_{2,\downarrow}^{\dagger} |0\rangle, \quad |\downarrow, \uparrow\rangle = \hat{c}_{2,\uparrow}^{\dagger} \hat{c}_{1,\downarrow}^{\dagger} |0\rangle, \quad (18)$$

and (complex) coefficients  $\alpha_i$ . For the ground state, one finds  $\alpha_1 = \alpha_2 \equiv \alpha_d$  and  $\alpha_4 = \alpha_3 \equiv -\alpha_s$  with a ground state energy  $E_0 = (U - \sqrt{U^2 + 16t^2})/2$ . In the non-interacting limit, these results reduce to  $\alpha_d = \alpha_s = 1/2$  and  $E_0 = -2t$ .

Any state of the form (17) can be written as a Gutzwiller wave function, no matter which form of the local correlation operator we choose. We demonstrate this for the correlation operator (3). In a general state  $|\Psi_0\rangle$ , we create two particles described by the operators

$$\hat{h}_{\sigma}^{\dagger} \equiv e^{i\phi_{\sigma}} u_{\sigma} \hat{c}_{1,\sigma}^{\dagger} + v_{\sigma} \hat{c}_{2,\sigma}^{\dagger} \quad (\sigma = \uparrow, \downarrow), \quad (19)$$

where  $v_\sigma \equiv \sqrt{1 - u_\sigma^2}$ . The variational parameters in (3) are also complex numbers in the following considerations and will be written as  $g_i = \bar{g}_i e^{i\kappa_i}$  with  $\bar{g}_i, \kappa_i \in \mathcal{R}$ . With this expression for  $g_i$  and Eq. (19), a general Gutzwiller wave function for the two-site Hubbard model has the form

$$|\Psi_G\rangle = \bar{g}_1 e^{i(\kappa_1 + \phi_\uparrow + \phi_\downarrow)} u_\uparrow u_\downarrow |d, \emptyset\rangle + \bar{g}_2 e^{i\kappa_2} v_\uparrow v_\downarrow |\emptyset, d\rangle + e^{i\phi_\uparrow} u_\uparrow v_\downarrow |\uparrow, \downarrow\rangle + e^{i\phi_\downarrow} v_\uparrow u_\downarrow |\downarrow, \uparrow\rangle. \quad (20)$$

We write the coefficients in (17) as  $\alpha_i = \bar{\alpha}_i e^{i\beta_i}$  and chose all phases such that  $u_\sigma, \bar{g}_i, \bar{\alpha}_i > 0$ . Then the comparison of the phases in (17) and (20) yields

$$\phi_\downarrow = \beta_4, \quad \phi_\uparrow = \beta_3, \quad \kappa_2 = \beta_2, \quad \kappa_1 = \beta_1 - \beta_3 - \beta_4. \quad (21)$$

Instead of normalising both states (17) and (20), it is easier (and equivalent) to set  $\bar{\alpha}_4 = 1$  and ensure the same for the corresponding coefficient in  $|\Psi_G\rangle$  by dividing (20) by  $v_\uparrow u_\downarrow$ . A comparison of the three remaining coefficients then leads to the equations

$$\bar{g}_1 u_\uparrow = \bar{\alpha}_1 v_\uparrow, \quad \bar{g}_2 v_\downarrow = \bar{\alpha}_2 u_\downarrow, \quad u_\uparrow v_\downarrow = \bar{\alpha}_3 u_\downarrow v_\uparrow, \quad (22)$$

which have the solution

$$\bar{g}_1 = \frac{\bar{\alpha}_1 \sqrt{1 - u_\downarrow^2}}{\bar{\alpha}_3 u_\downarrow}, \quad \bar{g}_2 = \frac{\bar{\alpha}_2 u_\downarrow}{\sqrt{1 - u_\downarrow^2}}, \quad u_\uparrow = \frac{\bar{\alpha}_3 u_\downarrow}{\sqrt{1 - u_\downarrow^2} (1 - \bar{\alpha}_3^2)}. \quad (23)$$

With this result, we have demonstrated that each state of the form (17) can be written as a Gutzwiller wave function with the correlation operator (3). The same can be shown for the other representations of the correlation operators in Sec. 2. Note that the parameters in the correlation operators are not uniquely defined by the correlated state. For example, any choice of  $u_\downarrow$  in (23) leads to the same Gutzwiller wave function after normalisation. This ambiguity will turn out to be the main obstacle in our application of the LFA, see below.

## 5. The TDGA for the two-site Hubbard model

As we will show in this section, one can gain valuable insight into the applicability of the time-dependent approach beyond the Gutzwiller approximation by a comparison with the exact results for a two-site Hubbard model. We shall discuss the low frequency (LFA) and fully time-dependent approach (FTDA) separately in the following two sections.

### 5.1. LFA

The LFA requires the calculation of the effective energy function (8). If we work, e.g., with the correlation operator (3) we need to minimise the energy with respect to  $g_1, g_2$  which gives us an effective energy  $E(u_\uparrow, u_\downarrow)$  as a function of the two parameters  $u_\sigma$  that determine  $|\Psi_0\rangle$ . This effective energy has then to be used in the equations of motion (10), (11). To demonstrate the substantial difficulties of

this approach we consider the case of pure charge fluctuations where

$$\bar{\alpha}_3 = \bar{\alpha}_4, \quad \beta_3 = \beta_4 = -\pi, \quad \beta_1 = \beta_2 = 0 \quad \text{in (17)}, \quad (24)$$

$$\phi_\uparrow = \phi_\downarrow = -\pi, \quad u_\uparrow = u_\downarrow \equiv u, \quad \kappa_i = 0 \quad \text{in (20)}. \quad (25)$$

In this case, the ground state (with  $\bar{\alpha}_3 = \bar{\alpha}_4 = 1, \bar{\alpha}_1 \equiv \bar{\alpha}_1^{\text{gs}}, \bar{\alpha}_2 \equiv \bar{\alpha}_2^{\text{gs}}$ ) is recovered for each value of  $u$  by setting  $\bar{g}_1^2 = (1 - u^2)/u^2 \bar{\alpha}_1^{\text{gs}}$  and  $\bar{g}_2^2 = u^2/(1 - u^2) \bar{\alpha}_2^{\text{gs}}$ , see Eqs. (23). As a consequence, the effective energy function (equivalent to the exact ground state energy),

$$E(\tilde{\rho}) = E(u_\uparrow, u_\downarrow) = E(u) = E_0 = (U - \sqrt{U^2 + 16t^2})/2, \quad (26)$$

is a constant, i.e., independent of the density matrix. This energy expression obviously leads to unphysical results if we evaluate the equations of motion (10), (11) and calculate, e.g., the charge susceptibility  $\chi_c(\omega) \equiv \langle\langle \hat{n}_1; \hat{n}_2 \rangle\rangle_\omega$  (with  $\hat{n}_i \equiv \hat{n}_{i,\uparrow} + \hat{n}_{i,\downarrow}$ ).

The problem with the correlation operator (3) stems from the fact that a fluctuation imposed on the state  $|\Psi_0\rangle$  can be fully reversed by a proper choice of the parameters  $g_1, g_2$ . Hence, the LFA does not even lead to the correct result in the non-interacting limit ( $U = 0$ ). Since (3) is a special version of the correlation operator (4), the latter runs into the same problem. Only if we add the transformation (5), the correlation operator (4) leads to a meaningful effective energy function  $E(\tilde{\rho})$  which, however, still does not give the exact response functions, see below.

The problems which we encounter with the correlation operators (3), (4) are, at first, surprising because the LFA has been applied successfully in a number of studies which were based on the energy function of the GA. Even a simple RPA calculation reproduces the correct response functions in the non-interacting limit. The reason why the GA leads to sensible results lies in a technical aspect of that approach whose physical consequences within the LFA have so far been overlooked. In order to evaluate expectation values in the limit of infinite dimensions analytically it is most convenient to work with the more general correlation operator (4) and to impose constraints which, for our two-site model, have the form

$$\langle \hat{n}_{i,\sigma} \rangle_{\Psi_0} = \langle \hat{n}_{i,\sigma} \rangle_{\Psi_G}. \quad (27)$$

As long as we are only interested in the variational ground state, these constraints do not change the physics, i.e., they just specify a particular representation of  $|\Psi_G\rangle$  within the variational freedom contained in the correlation operator (3). In the context of the LFA, however, the constraints have significant consequences because they ensure that fluctuations imposed on  $|\Psi_0\rangle$  are automatically also imposed on  $|\Psi_G\rangle$ . In this way, one avoids the difficulties which we observed for the correlation operator (3). An obvious remedy in our attempt to formulate the LFA for finite-dimensional systems (i.e., beyond the GA) is therefore to use these constraints here as well. Obeying the constraints, however, requires the use of the correlation operator (4) because with (3) (or (4) together with the transformation (5)) there are no variational parameters left in the minimisation (8).

We have calculated the charge susceptibility  $\chi_c(\omega)$  and the spin susceptibility  $\chi_s(\omega) \equiv \langle\langle \hat{S}_1^z; \hat{S}_2^z \rangle\rangle_\omega$  (with  $\hat{S}_i^z = (\hat{n}_{i,\uparrow} - \hat{n}_{i,\downarrow})/2$ ) with the three available LFA methods, i.e., we use the correlation operator (4) i) with the constraint (27), ii) with the transformation (5) where both i) and ii) are based on the exact evaluation of the energy functional. Finally, and iii), we use the correlation operator (4) and consider the energy function from the GA.



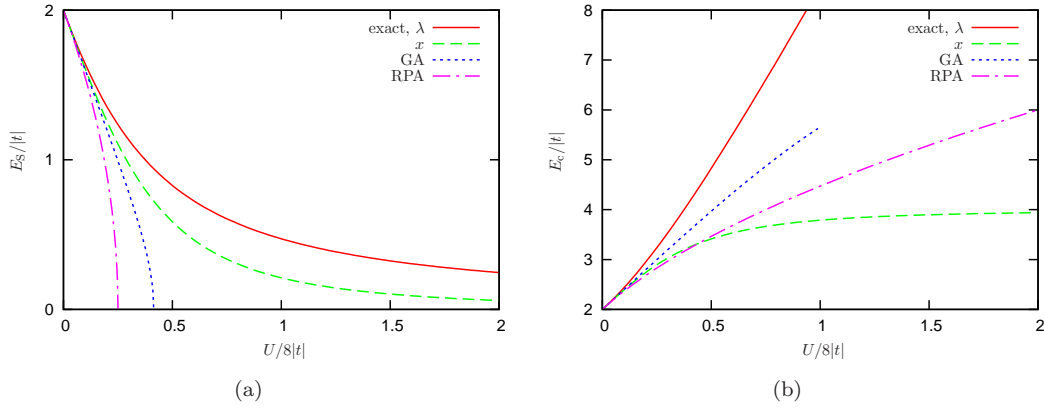


Figure 1. The excitation energies of the spin (a) and the charge susceptibility (b) calculated with: the operator (4) and the constraint (27) ( $\lambda$ ); the operator (4) and the transformation (5) ( $x$ ); the Gutzwiller approximation (GA), the random-phase approximation (RPA).

As in any finite system, the imaginary parts of  $\chi_{s/c}$  have  $\delta$ -like peaks at certain excitation energies of the system. It turns out that the exact solution of our two-site model has only one such energy for each of the two susceptibilities. These are

$$E_c^{\text{exact}} = (U + \sqrt{U^2 + 16t^2}) \quad \text{and} \quad E_s^{\text{exact}} = (-U + \sqrt{U^2 + 16t^2}). \quad (28)$$

In Fig. 1 we show these two excitation energies as well as their values in the Gutzwiller methods, mentioned above, and in the RPA (based on the Hartree–Fock approximation). The Gutzwiller method which employs the constraint (27) gives the exact results for the excitation energies. All other methods constitute approximations of varying accuracy.

In the GA and the RPA the excitation energies  $E_s$  go to zero at finite values of  $U$  ( $U_{\text{crit}}^{\text{RPA}} = 2t$ ,  $U_{\text{crit}}^{\text{GA}} = 8(\sqrt{2} - 1)t \approx 3.3t$ ). At these points, the GA and the Hartree–Fock energy function predict a spurious transition to an antiferromagnetic Néel state. This transition is absent if we work with the transformation (5) and the excitation energy is generally closer to the exact result for all  $U$  than those calculated with the two other methods. In contrast, the best approximation for the excitation energy  $E_c$  is the GA which, however, can only be applied up to the Brinkmann–Rice transition  $U = U_{\text{BR}} = 8|t|$  where the particles localise in that approach ( $\langle \hat{d}_i \rangle_{\Psi_G} = 0$ ).

In summary, the LFA forces us to work with the constraint (27) if we want to recover the exact results for the susceptibilities  $\chi_c(\omega)$  and  $\chi_s(\omega)$  of the two-site Hubbard model based on the exact evaluation of  $|\Psi_G\rangle$ . This prohibits us from using the transformation (5) which, however, is an indispensable element of our diagrammatic evaluation of expectation values beyond the GA. Therefore, we must conclude, that the LFA is only of limited use if we aim to improve our calculation of susceptibilities in finite dimensions.

## 5.2. FTDA

In Sec. 4, we have demonstrated that all states of the two-site Hubbard model can be written as a Gutzwiller wave function for each choice of the correlation operator. Therefore, a time-dependent Gutzwiller Ansatz in the Lagrangian (13) must reproduce the exact solution of the time-dependent Schrödinger equation for any time-dependent perturbation. This implies that for the susceptibilities which



derive from perturbations of the form (9) the exact results will also be recovered.

Note that the differential equations for the parameters  $z_i$  in the Gutzwiller wave function (e.g.,  $x_1, x_2$ ) are not necessarily linear and therefore much more complicated than the (linear) Schrödinger equation (e.g., for the coefficients  $\alpha_i$  in (17)). For the calculation of response functions, however, we will always expand the differential equations around the ground-state values of  $z_i$  to linear order and, in this way, end up with linear equations.

From the study of the differential equations for a two-site Hubbard model we do not learn much about the corresponding lattice problem, see below. Therefore, we conclude this section with the summary that all forms of correlation operators are, in principle, suitable for an improved calculation of two-particle excitation within the FTDA method. This holds, in particular, for the operator (4) with the transformation (5) which allows us to formulate an efficient diagrammatic evaluation of expectation values for finite-dimensional systems. In the following section, we will therefore derive the general FTDA equations for lattice systems.

## 6. The time-dependent Gutzwiller theory for finite dimensional systems

The transformation (5) ensures that

$$\hat{P}_i^\dagger \hat{P}_i = 1 + x_i \hat{d}_i^{\text{HF}} \quad , \quad \hat{d}_i^{\text{HF}} \equiv \hat{n}_{i,\uparrow}^{\text{HF}} \hat{n}_{i,\downarrow}^{\text{HF}} \quad , \quad \hat{n}_{i,\sigma}^{\text{HF}} \equiv \hat{n}_{i,\sigma} - n_{i,\sigma}^0 \quad (29)$$

is obeyed by the correlation operator (4). As we have shown in detail in Ref [2], this form of  $\hat{P}_i^\dagger \hat{P}_i$  allows for a very efficient diagrammatic expansion of expectation values. Based on the same diagrammatic expansion we will now derive the form of the Lagrangian (13) for our Gutzwiller wave functions. For all details on the diagrammatic method we refer the reader to Ref. [2].

A general time-dependent state  $|\Psi_0\rangle$  has the form

$$|\Psi_0(t)\rangle = \prod_{\gamma} [\hat{h}_{\gamma}^{\dagger}(t)]^{n_{\gamma}} |\text{vac}\rangle \quad \text{with} \quad \hat{h}_{\gamma}^{\dagger}(t) = \sum_{i,\sigma} u_{(i\sigma),\gamma}(t) \hat{c}_{i,\sigma}^{\dagger} \quad (30)$$

Here,  $n_{\gamma} \in \{0, 1\}$  determines which of the single particle states  $|\gamma(t)\rangle$ , described by the operators  $\hat{h}_{\gamma}^{\dagger}$  are occupied and  $u_{(i\sigma),\gamma}(t)$  is a (time-dependent) unitary transformation. The time dependent parameters  $\lambda_{i,\Gamma}(t)$  in (4) are written as

$$\lambda_{i,\Gamma}(t) = e^{i\varphi_{i,\Gamma}(t)} \sqrt{1 + \Theta_{i,\Gamma}(t) x_i(t)} \quad , \quad (31)$$

where  $\Theta_{i,\Gamma}(t)$  is given by the corresponding coefficients in (5), e.g.,  $\Theta_{i,\emptyset}(t) = n_{i,\uparrow}^0(t) n_{i,\downarrow}^0(t)$ . Note that the local expectation values  $n_{i,\sigma}^0$  are, like all elements of the non-interacting density matrix (7), given as functions of  $u_{(i\sigma),\gamma}(t)$ ,

$$n_{i,\sigma}^0(t) = \sum_{\gamma} n_{\gamma} |u_{i\sigma,\gamma}(t)|^2 \quad (32)$$

After having introduced all relevant time-dependent quantities, we will drop the explicit time dependence in the following considerations.

For the first term  $L^{(1)}$  in (13) we need to calculate

$$\langle \Psi_G | \dot{\Psi}_G \rangle = \langle \Psi_0 | \hat{P}_G^\dagger \dot{\hat{P}}_G | \Psi_0 \rangle + \langle \Psi_0 | \hat{P}_G^\dagger \hat{P}_G | \Psi_0 \rangle \quad (33)$$

The time derivative of  $|\Psi_0\rangle$  leads to

$$\frac{\langle\Psi_0|\hat{P}_G^\dagger\hat{P}_G|\dot{\Psi}_0\rangle}{\langle\Psi_G|\Psi_G\rangle} = \sum_{\gamma,\gamma',i,\sigma} n_\gamma \dot{u}_{i\sigma,\gamma} u_{i\sigma,\gamma'}^* \frac{\langle\Psi_0|\hat{P}_G^\dagger\hat{P}_G\hat{h}_\gamma^\dagger\hat{h}_\gamma|\Psi_0\rangle}{\langle\Psi_G|\Psi_G\rangle}. \quad (34)$$

Note that we cannot conclude at this stage that  $\gamma'$  has to be equal to  $\gamma$  in (34) as we could do in a time-dependent Hartree Fock calculation (with  $\hat{P}_G = 1$ ) or in the limit of infinite dimensions, see below. To evaluate (34) further we write it as

$$(34) = \sum_{\gamma,i,j,\sigma} n_\gamma \dot{u}_{i\sigma,\gamma} u_{j\sigma,\gamma}^* \frac{\langle\Psi_0|\hat{P}_G^\dagger\hat{P}_G\hat{c}_{i,\sigma}^\dagger\hat{c}_{j,\sigma}|\Psi_0\rangle}{\langle\Psi_G|\Psi_G\rangle} \equiv \sum_{\gamma,i,j,\sigma} n_\gamma \dot{u}_{i\sigma,\gamma} u_{j\sigma,\gamma}^* R_{(i\sigma),(j\sigma)} \quad (35)$$

where the expectation value  $R_{(i\sigma),(j\sigma)}$  can now be calculated by means of the diagrammatic method introduced in Ref. [2]. This leads to

$$R_{(i\sigma),(j\sigma)} = \delta_{i,j} n_{i,\sigma}^c + (1 - \delta_{i,j}) \left[ T_{i\sigma,j\sigma}^{(1),(1)} + (1 - n_{i,\sigma}^0) x_i T_{i\sigma,j\sigma}^{(1),(3)} - n_{j,\bar{\sigma}}^0 x_j T_{i\sigma,j\sigma}^{(3),(1)} - (1 - n_{i,\sigma}^0) n_{j,\bar{\sigma}}^0 x_i x_j T_{i\sigma,j\sigma}^{(3),(3)} \right] \quad (36)$$

where

$$n_{i,\sigma}^c = \langle\hat{n}_{i,\sigma}\rangle_{\Psi_G} = n_{i,\sigma}^0 + I_\sigma^{(2)} + x_i(1 - n_{i,\sigma}^0) I_{\bar{\sigma}}^{(2)} + x_i(1 - 2n_{i,\sigma}^0) I^{(4)} \quad (37)$$

is the (correlated) local particle number. Expressions for the diagrammatic sums in (36) and (37) have been derived in Ref. [2] and are given as

$$I_{[\sigma]}^{(4)[(2)]} \equiv \sum_k \frac{1}{k!} \sum_{l_1, \dots, l_k} \langle \hat{d}_i^{\text{HF}} [\hat{n}_{i,\sigma}^{\text{HF}}] \prod_{m=1}^k x_{l_m} \hat{d}_{l_m}^{\text{HF}} \rangle_{\Psi_0}^{\text{con}}, \quad (38)$$

$$T_{(i\sigma),(j\sigma)}^{(1)[(3)],(1)[(3)]}(k) \equiv \sum_k \frac{1}{k!} \sum_{l_1, \dots, l_k} \langle [\hat{n}_{i,\sigma}^{\text{HF}}] \hat{c}_{i,\sigma}^\dagger [\hat{n}_{j,\bar{\sigma}}^{\text{HF}}] \hat{c}_{j,\sigma} \prod_{m=1}^k x_{l_m} \hat{d}_{l_m}^{\text{HF}} \rangle_{\Psi_0}^{\text{con}}. \quad (39)$$

Here,  $\langle \dots \rangle_{\Psi_0}^{\text{con}}$  indicates that only connected diagrams are to be kept after the application of Wick's theorem. Note that, in the limit of infinite spatial dimensions (i.e., within the GA), we find  $n_{i,\sigma}^c = n_{i,\sigma}^0$  and the only non-zero diagram (39) is

$$T_{(i\sigma),(j\sigma)}^{(1)(1)} = \langle \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma} \rangle_{\Psi_0}. \quad (40)$$

With

$$\sum_j u_{j\sigma,\gamma}^* \langle \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma} \rangle_{\Psi_0} = u_{i\sigma,\gamma}^* \quad (41)$$

we then recover the result

$$\frac{\langle\Psi_0|\hat{P}_G^\dagger\hat{P}_G|\dot{\Psi}_0\rangle}{\langle\Psi_G|\Psi_G\rangle} = \sum_{\gamma,i,\sigma} n_\gamma \dot{u}_{i\sigma,\gamma} u_{i\sigma,\gamma}^* \quad (42)$$

which has been used in our previous work, Ref. [19].

For the second term in (33), we need to evaluate

$$\hat{P}_G^\dagger \dot{\hat{P}}_G = \sum_i \left( \prod_{j(\neq i)} \hat{P}_j^\dagger \hat{P}_j \right) \hat{P}_i^\dagger \dot{\hat{P}}_i \quad (43)$$

where

$$\hat{P}_i^\dagger \dot{\hat{P}}_i = \sum_\Gamma \left[ i\dot{\varphi}_{i,\Gamma} |\lambda_{i,\Gamma}|^2 \hat{m}_{i,\Gamma} - \frac{1}{2} (\dot{\Theta}_{i,\Gamma} x_i + \Theta_{i,\Gamma} \dot{x}_i) \hat{m}_{i,\Gamma} \right]. \quad (44)$$

Note that  $\Theta_{i,\Gamma}$  and  $x_i$  are real numbers and, therefore, the corresponding terms in (44) do not enter our real Lagrangian (13). Hence, its first part is given as

$$L^{(1)} = \sum_{i,j} \left[ \frac{i}{2} \sum_{\gamma,\sigma} n_\gamma (\dot{u}_{i\sigma,\gamma} u_{j\sigma,\gamma}^* R_{(i,\sigma),(j,\sigma)} - \text{h.c.}) - \delta_{i,j} \sum_\Gamma \dot{\varphi}_{i,\Gamma} m_{i,\Gamma} \right], \quad (45)$$

where we introduced the expectation value  $m_{i,\Gamma} \equiv \langle \hat{m}_{i,\Gamma} \rangle_{\Psi_G}$  which can be calculated diagrammatically. In infinite dimensions, as used in Ref. [19], it has the simple form  $m_{i,\Gamma} = |\lambda_{i,\Gamma}|^2 \langle \hat{m}_{i,\Gamma} \rangle_{\Psi_0}$ .

The Lagrangian (45) is complemented by the expectation value of the Hamiltonian in (13). Our diagrammatic approach leads to

$$L^{(2)} = - \sum_{i,j,\sigma} t_{i,j} \left[ q_{i,\sigma} q_{j,\sigma}^* T_{i\sigma,j\sigma}^{(1),(1)} + q_{i,\sigma} \alpha_{j,\sigma}^* T_{i\sigma,j\sigma}^{(1),(3)} + \alpha_{i,\sigma} q_{j,\sigma}^* T_{i\sigma,j\sigma}^{(3),(1)} + \alpha_{i,\sigma} \alpha_{j,\sigma}^* T_{i\sigma,j\sigma}^{(3),(3)} \right] - U \sum_i m_{i,d} \quad (46)$$

where

$$q_{i,\sigma} = \lambda_d^* \lambda_{\bar{\sigma}} n_{\bar{\sigma}}^0 + \lambda_\sigma^* \lambda_\emptyset (1 - n_{\bar{\sigma}}^0) \Big|_i = e^{-i\chi_\sigma} (q_{\emptyset,\sigma} + q_{d,\sigma} e^{-i\eta}) \Big|_i, \quad (47)$$

$$\alpha_{i,\sigma} = \lambda_d^* \lambda_{\bar{\sigma}} - \lambda_\sigma^* \lambda_\emptyset \Big|_i = e^{-i\chi_\sigma} (\alpha_{\emptyset,\sigma} - \alpha_{d,\sigma} e^{-i\eta}) \Big|_i, \quad (48)$$

and

$$q_{\emptyset,\sigma} \equiv |\lambda_d| |\lambda_{\bar{\sigma}}| n_{\bar{\sigma}}^0 \Big|_i, \quad q_{d,\sigma} \equiv |\lambda_\sigma| |\lambda_\emptyset| (1 - n_{\bar{\sigma}}^0) \Big|_i, \quad (49)$$

$$\alpha_{\emptyset,\sigma} \equiv |\lambda_d| |\lambda_{\bar{\sigma}}| \Big|_i, \quad \alpha_{d,\sigma} \equiv |\lambda_\sigma| |\lambda_\emptyset| \Big|_i, \quad (50)$$

$$\chi_\sigma \equiv \varphi_\sigma - \varphi_\emptyset \Big|_i, \quad \eta \equiv \varphi_\uparrow + \varphi_\downarrow - \varphi_\emptyset - \varphi_d \Big|_i. \quad (51)$$

As mentioned before, in infinite dimensions only the kinetic energy diagram (40) is non-zero. With this expression we recover the Lagrangian derived in [19]. As in that work, we have eliminated the phases  $\varphi_{i,\sigma}$ ,  $\varphi_{d,i}$  in favor of  $\chi_{i,\sigma}$  and  $\eta_{i,\sigma}$ . Note that after this elimination,  $\varphi_{\emptyset,i}$  does not appear anywhere in  $L^{(2)}$  and therefore can

be disregarded as a dynamical variable. The Lagrangian  $L^{(1)}$  then has the form

$$L^{(1)} = - \sum_i m_{i,d} \dot{\eta}_i - \sum_{i,\sigma} n_{i,\sigma}^c \dot{\chi}_{i,\sigma} + \frac{i}{2} \sum_{i,j,\sigma,\gamma} n_\gamma (\dot{u}_{i\sigma,\gamma} u_{j\sigma,\gamma}^* R_{(i,\sigma),(j,\sigma)} - \text{h.c.}) \quad (52)$$

$$- \sum_{i,j} \Omega_{i,j}(t) \left( \sum_\gamma u_{i,\gamma}^* u_{j,\gamma} - 1 \right)$$

where we have added a Lagrange-parameter term to ensure that  $u_{i,\gamma}$  is unitary.

With the Lagrangian (13) derived, it is now a straightforward task to set up the differential equations for our dynamical variables  $x_i$ ,  $u_{i\sigma,\gamma}$ ,  $\chi_{i,\sigma}$  and  $\eta_{i,\sigma}$ . If evaluated around the ground-state values of these properties, we are able to calculate two-particle response functions. Technically, the most challenging part is the calculation of first and second derivatives of the diagrams (38) and (39) with respect to the dynamical variables. Work on this numerical problem is in progress and will be published elsewhere.

## 7. Summary and Outlook

We have outlined a formalism which allows for the computation of collective excitations on top of the *exact* Gutzwiller ground state, i.e., beyond the Gutzwiller approximation corresponding to the limit of infinite spatial dimensions. We have outlined the approach by means of the two-site Hubbard model where it reproduces the exact excitation spectrum and have compared it with approximations used earlier in this context. In future work the method can be used in order to systematically improve the calculation of dynamical correlation functions based on the Gutzwiller wave-function. Especially interesting in this context is an investigation of the ‘Thouless criterion’ signaling the instability towards superconductivity since it is known [3, 14] that one has to go beyond the standard Gutzwiller approximation in order to stabilize SC order. Another obvious application is the study of excitations within the one-dimensional Hubbard model. It will then be interesting to see to which extend characteristic features of 1D correlated systems (e.g., spin-charge separation) are captured within the exact Gutzwiller correlations.

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